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Significant progress has been made in the following two research areas: I. Nonlinear spectroscopy of micrometer-sized multicomponent droplets; and II. Two- and three-dimensional scalar and velocity mapping.

I. Chemical species and physical properties of multicomponent liquid droplets in a spray combustor can be determined by a nonintrusive in-situ optical diagnostics techniques. A brief summary of the research accomplishments in the three areas related to the nonlinear optical interactions inside micron-sized droplets and the applications of such spectroscopy to determine the chemical and physical properties of the droplets: (1) Model for Nonlinear Optical Processes in Droplets, (2) Fluorescence Seeding of Stimulated Raman Scattering (SRS) of the Minority Species, and (3) Detection of Slight Shape Distortion by Spectroscopic Means. II. A review of the progress in our multi-dimensional scalar and velocity measurements in turbulent flames is also given. Among the accomplishments during the funding period are the following: (1) Scalar field measurements of differential diffusion effects in turbulent flows; (2) Development of a digital particle image velocimetry (PIV) technique for velocity field measurements in reacting and nonreacting flows; (3) Combination of the new digital PIV technique with previously developed fluorescence imaging techniques to allow simultaneous vector and scalar imaging in turbulent flames, and (4) Development of new mixture fraction imaging techniques for studying turbulent nonpremixed hydrocarbon flames.

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**FINAL REPORT**

**to the**

**Air Force Office of Scientific Research**

**Nonlinear Spectroscopy of Multicomponent Droplets**

**and**

**Two- and Three-Dimensional Measurements in Flames**

**AFOSR Grant No. 91-0150**

**February 1, 1991 - January 31, 1994**

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## INTRODUCTION

During the three years of AFOSR support, significant progress has been made in the following two research areas: (1) Nonlinear spectroscopy of micrometer-sized multicomponent droplets; and (2) Two- and three-dimensional scalar and velocity mapping. Specific details of our accomplishments regarding the use of nonlinear optical spectroscopy to characterize droplets and multidimensional diagnostics in flames can be found in the publications resulting from the research (See list starting on page 24). All these papers have been submitted to AFOSR in both preprint and reprint form.

## RESEARCH ACCOMPLISHMENTS

### I. Nonlinear Spectroscopy of Droplets

Nonintrusive in-situ optical diagnostics techniques have the potential of determining the chemical species and physical properties of multicomponent liquid droplets in a spray combustor. A spherical droplet or slightly deformed spherical droplet (in the form of an oblate or prolate spheroid) has the following unique characteristics: (1) enhances the internal intensity of the incident radiation; (2) increases the spontaneous emission rates for the lasing and the spontaneous Raman and Brillouin scattering processes; and (3) provides optical feedback for the internally generated nonlinear waves. Consequently, the input intensity threshold needed to achieve stimulated emission in the micron-sized droplets is much lower than that expected from a liquid in a micron-thick optical cell. Our research has shown that the nonlinear emission spectra from micron-sized droplets contain information on the chemical species, the species concentration, the droplet radius, and, more recently, the deformation amplitude of oblate and prolate spheroids.

Following is a brief summary of the research accomplishments in three areas related to the nonlinear optical interactions inside micron-sized droplets and the applications

of such nonlinear spectroscopy to determine the chemical and physical properties of the droplets:

**(1) Model for Nonlinear Optical Processes in Droplets**

We modified the standard one-dimensional coupled nonlinear-wave equations that are appropriate for stimulated Brillouin scattering (SBS) and stimulated Raman scattering (SRS) in an optical cell in order to accommodate the droplet geometry. To our knowledge, there does not exist a set of nonlinear-wave equations that can describe the experimental observations from single droplets, such as the time delays for the growth of SBS and SRS and the temporally correlated growths and decays among the SBS and the various-order Stokes SRS waves. In our heuristic model, the internal intensity spatial distribution is calculated with the well-established Lorenz-Mie formalism. The internal intensity temporal profile is the same as experimentally observed time profile. For each roundtrip around the droplet rim, the Raman (or Brillouin) wave experiences gain at the regions of high internal intensity and experiences loss all around the droplet rim because of radiation leakage, linear absorption, and intensity depletion associated with generating other nonlinear waves. The first-order Stokes SRS (or SBS) starts from the spontaneous Raman (or Brillouin) scattering. The higher-order Stokes SRS starts from both the spontaneous Raman scattering and the parametric signal associated with the four-wave mixing among the multi-order Stokes SRS waves.

There are only two adjustable parameters in our heuristic model. The first adjustable parameter is the Raman (or Brillouin) gain coefficient of a liquid. Because of the cavity QED effect, the gain coefficient in a droplet can be enhanced relative to that in an optical cell. The second adjustable parameter is the wave-vector mismatch  $\Delta k$  (or coherence length) among the nonlinear waves participating in the four-wave mixing process. The wave-vector mismatch among MDR's is difficult to predict because the MDR's can have different phase velocities that differ from the plane wave phase velocity

in a liquid. By adjusting the Raman (or Brillouin) gain coefficient and the wave-vector mismatch in our heuristic model, we were able to obtain good agreement with the experimentally observed temporal profiles of both the SBS and the first- through third-order Stokes SRS. The Raman and Brillouin gain coefficients affected the delay time between the appearance of the SBS and first-order SRS, after the pump laser is turned on. The wave-vector mismatch affected the temporal profiles of the SBS and the various order Stokes SRS. In addition, the wave-vector mismatch affected the correlation of the following: (1) the intensity valleys of the SBS with the intensity peaks of the first-order Stokes SRS; and (2) the intensity valleys of the first-order Stokes SRS with the intensity peaks of the second-order Stokes SRS. The adaptation of the standard nonlinear-wave equation of plane waves to accommodate the droplet geometry appears to be reasonably successful in explaining many observed nonlinear phenomena in droplets. (See publication #3.)

In diesel fuel droplets it was reported that SRS is not observable because the one- and two-photon absorption processes in the liquid suppress the SRS. One-photon absorption in the green can be minimized by selecting the input-laser wavelength ( $\lambda_{\text{input}}$ ) to be in the yellow or red region. Two-photon absorption in the blue may not be avoidable, for two reasons. First, the two-photon absorption bands of most liquids are not well known and are frequently ignored. Second, the two-photon absorption band in the blue is hard to avoid, because even if ( $\lambda_{\text{input}}$ ) is selected to be in the red region, ( $\lambda_{\text{input}}/2$ ) will be in the blue region. We incorporated two-photon absorption effects into our heuristic nonlinear wave-equation model in order to simulate the undesirable effect of some varying amounts of two-photon absorption.

Two-photon absorption can be grouped into two cases. The first is a degenerate two-photon absorption case involves two red-SRS photons that are distributed around the droplet rim. Another non-degenerate two-photon absorption case involves one red-SRS photon and one green pump-laser photon that is confined to a small focal region within

the droplet sphere. In the latter case, the non-degenerate two-photon absorption process competes with the SRS gain process. Amplification of the Raman waves can only occur when the Raman gain coefficient is larger than the 2-photon absorption coefficient. Thus, the non-degenerate two-photon absorption process is the main inhibitor of SRS in droplets. The degenerate two-photon absorption case, two-photon absorption only affects the steady-state SRS intensity, i.e., after the SRS threshold has been exceeded. A preliminary report of our study of two-photon effects was submitted as an abstract of a plenary lecture at the 1992 International Congress of Raman Scattering, held in Würzburg, Germany during August 31-September 4, 1992. (See publication #5.)

The most well-studied nonlinear processes in droplets are lasing, SBS, and SRS. The phase-matching condition is automatically satisfied for all these three processes. The other types of four-wave mixing (FWM) processes that have been observed in droplets are coherent anti-Stokes Raman scattering (CARS), coherent Raman gain, third-order sum frequency generation (TSFG), and stimulated anti-Stokes Raman scattering (SARS). SARS and CARS involve the same physical process. The CARS experiments require two external beams: one pump beam at  $\omega_{\text{pump}}$ ; and the other Stokes probe beam at  $\omega_{\text{Stokes}} = \omega_{\text{pump}} - \omega_{\text{vib}}$ , where  $\omega_{\text{vib}}$  is the frequency of the vibrational mode. By contrast, the SARS experiments require only one external pump beam at  $\omega_{\text{pump}}$ , because the Stokes probe radiation at  $\omega_{\text{Stokes}}$  is provided by the intense SRS generated within the droplet. Understanding SARS is important to the overall understanding of CARS in droplets. Our experimental observation on SARS in droplets demonstrated the importance of having the anti-Stokes frequency to be on or near an appropriate MDR. (See publication #4.) To our knowledge, there are no prior reports on the observation of SARS in droplets. In order to achieve the phase-matching angle between the pump and Stokes waves in the droplet, we focused the pump beam on the droplet edge and thereby, launched a pump wave with wave vectors that are more appropriate for phase matching with the Stokes SRS waves (circulating around the droplet rim) and the resultant SARS waves. From our experiments

with ethanol droplets, we noted that the first-order SARS intensity is  $10^4$ X lower than the first-order SRS intensity. The first-order SARS intensity is considerably larger than the second-order SARS intensity. The intensities of the second- and higher-order SARS were comparable, indicating that the first-order SARS process involves a resonant third-order susceptibility  $\chi^{(3)}$  and the higher-order SARS process involves a non-resonant  $\chi^{(3)}$ .

Our experimental results on TSFG in droplets clearly demonstrated the importance of having the sum frequency output be on or near an appropriate MDR, presumably because of wave-vector phase-matching considerations. (See publication #6.) For the FWM processes (such as TSFG and SARS) in an extended medium, wave-vector phase matching among the three generating waves and the resultant fourth wave is a consequence of spatial overlap consideration as the four waves co-propagate along the medium. There are two ways to envision the wave-vector phase-matching conditions in droplets. One approach is to decompose the standing-wave nature of a MDR as two counter propagating traveling waves that have equal but opposite phase velocity. The other approach for treating wave-vector phase matching in droplets is to calculate the spatial overlaps among the three generating waves and the resultant fourth wave, analogous to the case for wave-vector phase matching of FWM processes in an extended medium.

We have numerically calculated the spatial overlaps of various combinations of four MDR's associated with third-harmonic generation (THG) by the internal SRS waves that are on MDR's. The spatial overlaps among the four MDR's were calculated in the radial and in the two angular directions of the sphere. The treatment of phase matching of waves in droplets that are on MDR's is more accurate with the spatial overlap approach than with the phase velocity approach, which is only accurate for waves circulating in the equatorial plane. (See publications #7 and #11.)

A review of many of the nonlinear optical processes that can be observed with a single mode (injection-seeded laser) is presented at a SPIE meeting in Dallas, Texas.



(See publication #2.) An extensive manuscript will soon be published in the conference proceedings of 5th International Topsøe Summer School on Nonlinear Optics, held in Aalborg, Denmark, 3-8 August, 1992. (See publication #16.)

## **(2) Fluorescence Seeding of Stimulated Raman Scattering (SRS) of the Minority Species**

We have been investigating ways to improve the generation of SRS for the minor species (benzene, an example of a carbon ring compound which give rise to soot) in the presence of the major species (dodecane). In a multicomponent fuel droplet, one component can evaporate faster than the other components. An example is a low boiling point liquid (pentane) mixed with a high boiling point liquid (1-methyl-naphthalene). Consequently, the detection by SRS of the low boiling point component, after significant amount of pentane vaporization has taken place, can be a problem. (See publication #12.) The input intensity needed for reaching the SRS threshold for the minor species (pentane in this example) can not be achieved, because the generation of SRS for the major species (1-methyl-naphthalene in this example) can deplete the internal input intensity, making the latter insufficient to pump the minor species.

For species detection of a multi-component fluid droplet by SRS detection, fluorescence seeding can preferentially increase the SRS intensity of the minor species. The fluorescence spectrum is independent of the pump wavelength. By contrast, the Raman spectrum is dependent on the pump wavelength, because the Stokes Raman frequency shift is always equal to the vibrational frequency of the molecule. Good spectral overlap between the fluorescence and the Raman scattering of the minor species is required for selective fluorescence seeding of the SRS of the minor species. In addition, it is required that the Raman scattering of the major species must be outside the fluorescence emission profile.

Conventionally, SRS builds up from spontaneous Raman noise. In the absence of injected external seeding radiation at the Stokes wavelength, the SRS processes can be

considered as the amplification of spontaneous Raman noise by as much as  $e^{30}$ . However, when the droplet contains dye molecules that fluoresce within the Stokes Raman wavelength region of the minor species, the Stokes SRS for the minor species no longer needs to start only from spontaneous Raman noise. The SRS intensity is allowed to build up from a much larger initial value, that is determined by the fluorescence of the dye molecules rather than from the weaker spontaneous Raman intensity. Even though the typical dye concentration is extremely low (e.g.,  $10^{-7}$  to  $10^{-6}$  M), the fluorescence noise is still much larger than the spontaneous Raman noise, because the fluorescence cross section ( $\sigma = 10^{-16}$  cm<sup>2</sup>) is fourteen orders of magnitude larger than the spontaneous Raman cross section ( $\sigma = 10^{-30}$  cm<sup>2</sup>) of the minor species. Fluorescence seeding effectively lowers the input-pump laser intensity needed to achieve a certain detectable SRS intensity of the minor species. Moreover, fluorescence seeding also increases the growth rate of the SRS intensity, because the parametric growth rate is dependent on the product of the input-pump intensity and the Stokes intensity that has contributions from both the Raman and fluorescence emissions. (See publication #8.)

However, we have recently concluded that, accompanying the fluorescence seeding, there exist some increase of the SRS gain associated with the dye-lasing gain. We are finishing a series of experiments to further our understanding of the roles of fluorescence seeding and the concomitant "extra gain" provided by the population-inverted dye molecules. We have demonstrated that we can achieve the improved sensitivity of SRS of a minor species (3% benzene) in dodecane droplet containing  $2 \times 10^{-6}$  M R560 dye. (See publication #15.) Without the addition of the R560 dye or not having the SRS wavelength of benzene overlap spectrally with the fluorescence/lasing band of R560, we were unable to detect the SRS signal from 3% benzene in dodecane droplets. Without adding R560, the lowest benzene concentration we can detect is 5% in dodecane.

### (3) Detection of Slight Shape Distortion by Spectroscopic Means

In the past, we have "assumed" the shape of our droplets to be spherical. The special locations of MDR's are described exactly by the Lorenz-Mie theory that is appropriate for electromagnetic waves interacting with dielectric objects with spherical symmetry. MDRs are the natural modes of the droplet and are labeled by three indices: (1) mode order  $\ell$ , which specifies the number of intensity peaks as the radial distance is increased from the center to the droplet surface; (2) mode number  $n$ , which specifies the number of intensity peaks in the equatorial plane as the azimuthal angle  $\phi$  is varied from  $0^\circ$  to  $180^\circ$ ; and (3) azimuthal modes number  $m$ , which can assume the values  $m = \pm n, \pm(n-1), \dots, 0$ . For a perfect sphere, all the azimuthal modes (with different  $m$  values) for a fixed  $n$  have the same frequencies. The azimuthal modes are, therefore,  $2(n+1)$  degenerate. However, for droplets with small amounts of shape distortions, the degeneracy of the azimuthal modes are partially lifted. For MDR's with a particular  $\ell$  and  $n$ , the spectral frequencies of the MDR's with different  $m$  values are as follows:

$$\omega(m) = \omega_0 \left[ 1 - \frac{e}{6} \left[ 1 - \frac{3m^2}{(n+1)} \right] \right]$$

where  $\omega_0$  is the frequency of the degenerate MDR for a perfect sphere. The magnitude of the shape distortion  $e \equiv (r_p - r_e)/a$ , where  $r_p$  is the polar radius,  $r_e$  is the equatorial radius, and  $a$  is the radius of the equivolume sphere. Note that the frequency shift from  $\omega_0$  is dependent on  $m^2$  and on the sign of  $e$ . For an oblate spheroid,  $r_p < r_e$  and, therefore,  $e$  is negative. For a prolate spheroid,  $r_p > r_e$  and, therefore,  $e$  is positive. Thus by measuring the frequency shifts of the various MDR's with different  $m$  values, the absolute droplet shape (whether it is prolate or oblate) and the distortion amplitude  $e$  can be deduced spectroscopically.

We have observed the frequency splitting of degenerate spherical cavity mode by using a Fabry-Perot interferometer to analyze spectrally the SRS emerging from a single droplet, which is flowing in a continuous stream. Because of the inertial effect, the droplet shape is distorted into a prolate spheroid, with the axis of symmetry (z-axis containing the droplet poles) along the flow direction. Unlike a sphere, where all the great circles have the same circumference, the great ellipses of a prolate spheroid do not have the same length. For an oblate spheroid, the round trip distance is slightly larger around the droplet equator. Hence, the morphology-dependent resonance (MDR) of the droplet at the equator shifts to a longer wavelength. Furthermore, for an oblate spheroid, the round trip distance is slightly smaller around the droplet poles. Hence, the MDR of the droplet at the poles shifts to a shorter wavelength. The wavelength positions of the MDRs with different inclination angle  $\theta$  from the droplet poles are explicitly given from the perturbation theory developed by the group at the Chinese University of Hong Kong (See equation above.) In between the equator and the pole, the MDRs appear at discrete wavelengths. We have deduced the droplet shape distortion to be one part in  $10^3$  by fitting the observed discrete SRS peaks, corresponding to MDR's with different  $m$  values. In specific, for MDR's with  $m = n$ ,  $m = (n - 2)$ ,  $m = (n - 4)$  and  $m = (n - 6)$ . (See publication #1.)

We have also observed that the SRS (on a MDR inclined at  $\theta$ ) will precess in time. From the ray-optics view point, the radiation that circulates once around the oblate droplet rim or around a great ellipse does not begin and end at the same point. The end point is shifted by a small angle  $d\phi$  because of the shape distortion. After many trips around the droplet or after one precession period, the radiation will return to an inclination of  $\theta$ .

We were able to measure the precession period of the SRS that is on a MDR inclined at angle  $\theta$  using a streak camera. The relationship  $m = n \cos\theta$  is well known in angular momentum theory in quantum mechanics. With the help of Professor Kenneth Young of the Chinese University of Hong Kong, and by using the perturbation theory for MDR shifts as a function of  $\theta$ , we were able to deduce the droplet shape distortion from the precession frequency. The droplet shape distortion was deduced to be 7 parts out of  $10^3$ . The discrepancy between the previous frequency-splitting measurement (of 1 part out of  $10^3$ ) and the present precession-time measurement (of 7 parts out of  $10^3$ ) was attributed to the following experimental parameters: (1) the droplets were larger for the time-dependent experiment; and (2) the droplets were falling faster in the time-dependent experiment. Because the shape distortion is highly dependent on both the Weber and Reynolds numbers, we were satisfied that the time measurements gave a larger distortion than the frequency measurements. (See publications #9 and #10.)

The frequency splitting of MDR's with different  $m$  values (as observed in the frequency domain with a Fabry-Perot interferometer) and the precession frequency of a particular group of MDRs with values around  $m$  (as observed in the time domain with a streak camera) were reported in a SPIE conference in Shanghai, China. (See publication #10.) Both the frequency splitting and precession frequency can not provide the sign of the distortion amplitude, i.e., whether the droplet is prolate or oblate. A totally new technique was developed for the sole purpose of determining the sign of  $e$ .

In 1984 in order to determine the evaporation of droplets, we used the frequency shift in the lasing spectrum from individual droplets in a continuous stream, [for details see H.-M. Tzeng, K. F. Wall, M. B. Long, and R. K. Chang, "Evaporation and Condensation Rates of Liquid Droplets Deduced from Structure Resonances in the Fluorescence Spectra," *Opt. Lett.* **9**, 273 (1984)]. Since then, we were inspired by the theoretical papers of Prof. William Sirignano on three interacting droplets that exhibit different evaporation rates.

In the calculation, the lead droplet has the largest evaporation rate and the third droplet has nearly the same evaporation rate of a continuous stream having the same droplet-droplet separation.

In order to make contact with the theoretical models, we initiated an experiment to measure the evaporation rate of closely spaced droplets in a segmented stream. A pair of constant-voltage electrostatic-deflection plates were used to deflect the charged droplets in an undesired segment. The uncharged droplets in the desired segment will remain undeflected. Because of induced charges and the larger drag coefficients for the lead droplet, the second droplet coalesces with the lead droplet, making the new lead droplet  $(2)^{1/3}$  or  $(3)^{1/3}$  times larger than the trailing droplets. We attempted to use a charge-compensation technique (developed for ink-jet printers) as a means of producing a lead droplet of equal radius with respect to the trailing droplets. However, we concluded that the larger drag coefficient of the lead droplets will always cause coalescence with the second trailing droplet.

In spite of the fact that our lead droplet is larger than the trailing droplets, we noticed several interesting features regarding the evaporation rates of individual droplets: (1) the lead droplet evaporates faster than all the trailing droplets; (2) each droplet evaporates a little faster than its trailing droplet; and (3) only by the 6th trailing droplet, does the evaporation rate equal the evaporation rate of a continuous stream. (See publications #13 & #17.)

While studying the evaporation rates of closely interacting droplets, we were amazed that the spectral resolution of a high-resolution spectrograph (not an interferometer) were sufficient for us to resolve the frequency shifts of the lasing emission emerging from different parts of the droplet rim. The CCD camera, that was placed at the exit plane of the spectrograph was able to provide spatial and spectral information of the lasing emission

from different parts of the droplet rim. The spatial and spectral data had the form of  $\supset$ -shaped curve or  $\subset$ -shaped curve, depending whether the droplet shape is prolate or oblate. Thus, for the first time, we were able to deduce the absolute sign of  $e$  from the frequency shift associated with deformed droplets. From the curvature of the  $\supset$ -shaped or  $\subset$ -shaped curve, we were able to deduce the sign and the magnitude of  $e$ . (See publication #14.) Finally, we are in the position to measure the droplet evaporation rate, shaped distortion amplitude, the distortion shape, and the relative drag coefficient among closely interacting droplets.

## II. Two- and Three-Dimensional Measurements in Flames

Laser diagnostic techniques have been developed that are capable of two- and three-dimensional mapping of scalars and velocities in turbulent flames. Whenever possible, the techniques are tailored to measure quantities and flow configurations of current interest to combustion modelers. The availability of quantitative data on the spatial and temporal characteristics of structures in turbulent reacting flows will aid in understanding the interaction of chemical reactions with the turbulent motion. A better understanding of this key interaction is important for testing existing models of turbulent combustion as well as for suggesting new models. In the following sections, some of the major accomplishments of the three-year funding period are outlined.

### (1) Scalar Measurements in Reacting and Nonreacting Flows

With AFOSR funding, we have developed a number of new laser-based imaging techniques for measuring scalars in turbulent flows and flames (e.g., see publications #18 and #19.) After the initial development phase, it is our goal to continue refining the techniques and, in collaboration with other combustion researchers, to apply these techniques to the study of relevant problems (See publication #20.) One example of such a study is the simultaneous use of Lorenz-Mie and fluorescence imaging to study

differential diffusion effects in a turbulent nonreacting jet. In the experiment, the use of two different scattering mechanisms allowed the measurement of the difference between the concentration profiles of a nondiffusive marker (aerosols) and a diffusive one (biacetyl). During the first year of the grant, analysis was performed on data obtained in collaboration with researchers from the University of Sydney. Matched instantaneous image pairs of biacetyl and particle concentration showed differences; the particle images exhibited sharper contours and a more convoluted structure with finer detail. The fluorescence images showed a "superlayer" similar to that found in Rayleigh images in flows of this kind. Subtraction of one normalized image from the other in each pair revealed numerically greater overall diffusion of the biacetyl gas, a direct indication of its molecular contribution to total mixing. These findings support the argument that molecular diffusivity in general, and differential diffusion between species in particular, must be accounted for in the interpretation of laboratory flame results that are used to validate and test combustion models. (See publications #21 and #22.)

## **(2) Digital Particle Image Velocimetry**

Particle image velocimetry (PIV) has been established as a viable means for making quantitative measurements of two velocity components within a plane intersecting a flow. The technique is based on recording images of a seeded flow illuminated by a multiple-pulsed laser sheet. Two components of the velocity are determined from the separation of the particle images and the known time between laser pulses. In most of the work done to date, photographic film is used to record the images because of its high spatial resolution. In the last few years, however, electronic imaging devices with high spatial resolution have become available. We have investigated the use of a cooled 2048 x 2048 pixel CCD detector for recording PIV data.

There are several advantages to the use of electronic imaging for PIV. Probably the most significant is the elimination of the need for photographic processing of the film. The data are stored directly in the computer and so the human intervention



required in developing and digitizing of photographic negatives is eliminated. In addition, the images are available in nearly real time allowing on-line optimization of the focusing, temporal separation of the laser pulses, seeding density, and illumination sheet intensity. An additional advantage of using cooled CCD arrays relates to their large linear dynamic range. By taking advantage of this characteristic, we have successfully determined velocity fields in flows with significant variations in seeding density (flames are generally in this category). This capability is also useful in resolution of the directional ambiguity in PIV.

Experimentally, the use of a cooled CCD array for digital PIV is quite straightforward. The second or third harmonic of a double-pulsed Nd-YAG laser is formed into an illumination sheet that intersects the flow. The laser pulse separation can be varied between 40 and 200  $\mu\text{sec}$ , and is selected according to the mean flow velocity. The scattered light is imaged onto the CCD detector with the optical axis of the collection lenses oriented normal to the illumination sheet. Subsets of the large image can be directly analyzed by numerical Fourier methods to yield the autocorrelation of the sub-image, which is in essence the local velocity. By processing sub-images over the entire image, the velocity field can be obtained. Our current CCD detector allows resolution of velocities on a 32 x 32 element grid, with a spatial resolution for each velocity vector on the order of 0.3 mm.

Initial experiments in both nonreacting and reacting flows have been performed. For the non reacting case, the flow observed was an air jet seeded with submicron particles from an atomizing aerosol generator. Successful PIV measurements were also made in an alumina-seeded premixed methane-air flame.

### (3) Simultaneous Velocity and Scalar Imaging

Another advantage of the digital PIV technique described above is its compatibility with previously developed scalar imaging techniques. Although several researchers have performed simultaneous velocity and scalar measurements at a single

point, the capability of obtaining velocity and scalar images is new. In an initial set of experiments, laser-induced biacetyl fluorescence has been combined with digital PIV to allow simultaneous measurement of the nozzle gas concentration and the velocity field in a turbulent nonreacting jet. The nozzle gas was seeded with biacetyl as a marker, and both the nozzle gas and the coflow were seeded with submicron-sized aerosols for the PIV measurements. The third harmonic of a Nd-YAG laser (355 nm) excited biacetyl fluorescence at 470 nm, which was imaged onto a 384 x 576 pixel CCD detector through an interference filter. The Lorenz-Mie scattering from the aerosols was imaged onto a 2048 x 2048 element CCD for the velocity measurement.

More recently, the velocity and scalar imaging work has been extended to reacting flows. In these experiments, biacetyl fluorescence and digital PIV were used to map the unburned gas concentration and the velocity in a turbulent premixed methane-air flame. Simultaneous concentration and velocity data in flames show the position of the flame front as well as the velocity of the gases through the flame front. These data should be useful for studying extinction and for understanding the coupling between the turbulence and the chemical reactions. (See publication #23).

#### (4) Two-Dimensional Measurements of the Mixture Fraction

With AFOSR funding, we have initiated a set of experiments in which we hope to develop a new imaging technique for determining the mixture fraction; a quantity directly relevant to modeling nonpremixed turbulent combustion. The availability of mixture-fraction images in flames will be an important step in resolving issues related to the scales of flame structure and the operative burning regimes within turbulent nonpremixed flames.

The mixture fraction,  $\xi$ , (defined as the mass fraction of atoms that originated from the fuel stream) is of critical importance for modelers of turbulent nonpremixed combustion. Since this conserved scalar quantity is not affected by the chemical

reaction, it gives valuable information on the mixing process. Under certain conditions, (high Damkohler number, chemical equilibrium, equal diffusivities) this conserved scalar can be used to derive essentially all of the quantities of interest in the flame. Experimentally, the determination of the mixture fraction in nonpremixed flames is quite difficult since it involves simultaneous monitoring of a large number of species. At Sandia Laboratory the mixture fraction has been determined at a single point from measurements of all major species using spontaneous Raman scattering. Results from these studies are among the most complete sets of experimental data available in turbulent flames. The data from single-point measurements are incomplete, however, because of the lack of multi-dimensional information required to obtain gradients. The scalar dissipation,  $\chi$  (defined as  $\chi = 2D \nabla \xi \cdot \nabla \xi$  with  $D$  the diffusivity), determines the rate of molecular mixing and is widely used in modeling turbulent reacting flows. Therefore, techniques capable of two-dimensional or preferably three-dimensional measurements of the mixture fraction are needed.

Because the scalar dissipation is calculated from the square of the mixture fraction gradient, the scalar dissipation results are extremely sensitive to noise in the experimental data. Therefore, it is critical to obtain mixture fraction images with high signal-to-noise ratio. Because of the weakness of the Raman process, prospects for extending the single-point Raman measurement of the mixture fraction to two dimensions are remote. A means of determining the mixture fraction without measuring all of the major species has recently been proposed by Stårner et al. [More detail is available in S.H. Stårner, R.W. Bilger, R.W. Dibble, and R.S. Barlow, "Measurements of Conserved Scalars in Turbulent Diffusion Flames," *Combust. Sci. Tech.* **86**, (1992) p. 223]. In this work it was shown that the mixture fraction can be determined in three different ways from simultaneous measurement of only two quantities. The validity of the method requires a unity Lewis number and a one-step reaction between fuel and oxidizer. The measured quantities are used to form

a conserved scalar from which the mixture fraction is determined in an iterative process. We have performed a number of experiments in which the mixture fraction was obtained from images of the Rayleigh scattering and the fuel concentration. These experiments were conducted in our laboratory with the collaboration of researchers at the University of Sydney.

The flame selected for our first study was a turbulent nonpremixed acetaldehyde flame. Acetaldehyde ( $\text{CH}_3\text{CHO}$ ) was chosen for its relatively high fluorescence yield and small variation of fluorescence intensity with temperature, which allows the fuel concentration to be found directly from the acetaldehyde fluorescence. An alternative means of measuring fuel concentration is Raman imaging, but because of the need for high signal-to-noise ratio to determine scalar dissipation, the more efficient fluorescence process was investigated first.

In our Rayleigh scattering/acetaldehyde fluorescence imaging experiments, the flow was illuminated by two overlapping laser sheets. The first, formed from the second harmonic of a Nd:YAG pumped dye laser (320 nm, 10 mJ per 10 ns pulse, 10 mm sheet height), excited fluorescence from the acetaldehyde fuel. The second illumination sheet, from a flashlamp-pumped dye laser (532 nm, 2 J per 2  $\mu\text{s}$  pulse, 2 cm sheet height), excited Rayleigh scattering. The fuel fluorescence was imaged onto a gated-image intensifier that was optically coupled to a cooled CCD detector. The Rayleigh scattering was imaged onto a separately gated intensifier optically coupled to the second CCD detector. The two lasers and image intensifiers were fired sequentially with a pulse separation of  $\sim 3 \mu\text{sec}$ . This temporal separation ensured that there was no interference between the detected signals from the two laser sheets, but was short compared to the smallest flow time scales.

The raw images were corrected for background and non uniform detector response. In addition, since the magnification of the two images and the illumination sheet size were different, the necessary rotation, translation, scaling, and cropping

were applied to the images to allow them to be compared on a pixel-by-pixel basis. The fuel was a mixture of acetaldehyde diluted 1/1 by air (on a mass basis) to eliminate soot. It emanated from a piloted burner ( $d = 3.8$  mm) with a velocity corresponding to  $Re = 18,000$ . The flame was enshrouded in a low-velocity filtered-air coflow in order to keep the measurement area clear of particles, which would interfere with the Rayleigh images.

A conserved scalar,  $\beta$ , based on the fuel mass fraction ( $Y_f$ ) and the enthalpy ( $H = c_p T/Q$ ) served as the basis for determining the mixture fraction:

$$\beta = Y_f + c_p T/Q$$

$$\xi = \frac{\beta - \beta_2}{\beta_1 - \beta_2} = \frac{Y_f + c_p/Q(T-T_2)}{Y_{f,1} + c_p/Q(T_1-T_2)}$$

where the subscripts denote the fuel (1) and oxidizer (2) streams.  $Y_f$  is obtained from the fluorescence images and  $T$  is related to the Rayleigh images. The mixture fraction can be expressed in terms of the fluorescence signal,  $F$ , and the Rayleigh signal,  $Ra$ , as follows:

$$\xi = \frac{C_1 a_T F}{W Ra F_{cal}} + C_2 (a_T/Ra - T_{ambient})$$

where  $C_1$ ,  $C_2$ , and  $F_{cal}$  are calibration constants,  $a_T$  is proportional to the local Rayleigh cross section, and  $W$  is the local mixture molecular weight. Since  $a_T$  and  $W$  are both functions of  $\xi$ , an iterative process was used on a pixel by pixel basis to derive the temperature and mixture fraction. The procedure converges quickly, usually within fewer than four iterations.

Preliminary data on the calculated temperature and mixture fraction obtained from the instantaneous Rayleigh/fluorescence images show several interesting features. Evident in the temperature mappings are the high temperature zones on the outer edge of the flame. As expected, the mixture fraction peaks in the unburned regions near

the center of the jet. Results of these early experiments were presented at several international meetings. (See publications #24 and #25.)

A closer inspection of the mixture fraction images obtained from the acetaldehyde flame shows a slight decrease in the mixture fraction on the rich side of the stoichiometric value, where the temperature has not yet peaked. A similar dip in the mixture fraction on the rich side of the flame front was noted in measurements performed in a laminar flame. One explanation of this behavior is pyrolysis of the acetaldehyde fuel. To investigate this, experiments were done in which the acetaldehyde fluorescence and fuel Raman scattering were measured simultaneously. The C-H Raman scattering should be relatively insensitive to the breakup of the acetaldehyde into other hydrocarbon fragments. The fluorescence, on the other hand, is expected only from the acetaldehyde molecule itself, so differences in the Raman and fluorescence traces may be indicative of pyrolysis. The experimental configuration was nearly the same as for the fluorescence/Rayleigh experiments. Two changes were required: first, the appropriate interference filter was added in front of the detector used to detect the Raman scattering, and second, the flashlamp-pumped dye laser was focused to a line rather than a sheet to provide increased Raman signal. A comparison was made of acetaldehyde fluorescence and C-H Raman intensity along lines intersecting laminar and turbulent flames. In both cases, the agreement was quite good despite the relatively noisy Raman signal. These results support the assertion that acetaldehyde fluorescence is a good alternative to Raman scattering as a means of marking the fuel, but do not explain the anomalous dip in the mixture fraction on the fuel rich side of the stoichiometric value.

In a recent collaboration, Peter Lindstedt of Imperial College performed a detailed calculation of acetaldehyde chemistry in an opposed jet configuration (See publication #26). The results shed considerable light on the mixture fraction obtained in our experiments. Calculation of the mixture fraction from the computed temperature and fuel mass fraction using the two-scalar scheme outlined above results in a dip in mixture

fraction on the rich side of the flame similar to those obtained in the experiments. One of the most striking features of the calculation is the high concentration of CO (a maximum mole fraction of nearly 20%) present in the flame. The combustion characteristics of acetaldehyde appear to differ significantly from that of methane, so that the two scalar approach that worked well for methane and propane may not work as well for acetaldehyde. As one way to check that the results of the calculation are in agreement with the experimental data, the computed results were used to generate plots of the expected Raman and fluorescence signals. The results were in good qualitative agreement with our experiments.

Another technique that has been proposed as a way of marking the fuel in hydrocarbon flames is to introduce acetone as a tracer and detect acetone fluorescence. Acetone is easily seeded into flows and has only mild toxicity. However, an unresolved issue related to the use of acetone as a fuel tracer in flames relates to the behavior of the fluorescence as a function of temperature. To test the use of acetone as a fuel tracer for determination of mixture fraction, the fluorescence/Raman setup described above was used to investigate laminar and turbulent acetone-seeded methane flames.

The acetone was seeded into the methane fuel by bubbling methane through liquid acetone heated to 30° C. The methane and acetone vapor were diluted with air giving a final mixture of 70% air, 25% methane, and 5% acetone by volume. The laminar flame was stabilized on a 16 mm diameter nozzle, and the gas mixture had a Reynolds number of 700 at the nozzle exit. Results of the fluorescence/Raman comparison in the laminar flame show a clear departure between the normalized fluorescence and Raman signals, with the fluorescence signal exceeding the Raman signal in the fuel-rich region of the flame. This behavior is consistent with the fluorescence increasing as a function of temperature as reported by other researchers (e.g., see Tait, N.P., and Greenhalgh, D.A., *Twenty-Fourth Symposium (International) on Combustion*, The Combustion Institute, 1992, p. 1621). A comparison of the normalized Raman and fluorescence

signals in a turbulent flame showed similar results, with the fluorescence signal greater than the Raman signal in some locations. A paper describing the various two-scalar techniques that we have investigated for mixture fraction imaging has been accepted for presentation at the Twenty Fifth International Symposium on Combustion in Irvine CA in July 1994. (See publication #27.)

To avoid the problems related to the use of acetaldehyde or acetone as the fuel, a set of experiments were done in which the concentration of fuel was inferred from the Raman scattering of the vibrational C-H stretch of methane. The main drawback to Raman scattering is the extremely small Raman scattering cross section and, consequently, the difficulty in getting sufficient signals. As early as 1985, we demonstrated simultaneous Raman and Rayleigh imaging, although the data were not used to determine mixture fraction. Previous Raman imaging experiments made use of a multipass optical cell to form the illumination sheet. In this work, a multipass cell was not used, but a two-pass arrangement allowed two components of the scalar gradient to be determined along a line.

In the Raman and Rayleigh line imaging measurements, a flashlamp-pumped dye laser provided up to 1 J of energy at 532 nm. To reduce the beam divergence of the laser and thereby reduce the waist of the focused beam, the laser cavity was lengthened to 3.1 m. The beam was focused into the flame initially by a spherical lens. After passing through the flame, the beam was re-collimated by a second lens, reflected from a planar mirror, and refocused into the flame. The two beams were aligned so that the reflected beam passed just over the top of the incoming beam. This two-beam approach allowed gradients to be obtained in both the radial and axial directions. The beam waist was measured to be 0.6 mm.

The scattered Raman and Rayleigh light was detected by two intensified CCD detectors oriented perpendicular to the laser beam. The Raman-scattered light was isolated by an interference filter and focused onto a single-stage image intensifier



by an f1.2 camera lens. The output of the intensifier was optically coupled to a cooled CCD detector. An f4.0 camera lens imaged the Rayleigh scattering onto a two-stage image intensifier, which was optically coupled to the second cooled CCD detector. Both image intensifiers were gated coincident with the laser pulse to eliminate flame luminosity. (Note that the use of the intensifiers was primarily for gating rather than for high gain.) The digitized output from both cameras was read into a single lab computer.

The raw Raman and Rayleigh line images were corrected for background and nonuniform detector response. In addition, since the magnification of the two images was slightly different, the necessary rotation, translation, scaling, and cropping were applied to the images to allow them to be compared on a pixel-by-pixel basis.

The volume corresponding to each pixel was  $.09 \times .09 \times 0.6 \text{ mm}^3$ , with the largest value corresponding to the beam thickness. Since the beam thickness represents the main limitation to the spatial resolution in these measurements, the signals were integrated in the cross beam direction and 3 pixel smoothing was applied in the radial direction. The resulting images consist of two lines radially across the flow corresponding to the main and reflected beams with a pixel resolution of  $0.27 \times 0.45 \times 0.6 \text{ mm}^3$ .

The flame investigated was an axisymmetric, piloted methane-air flame. The main fuel nozzle had a diameter of 3.8 mm. The main jet was surrounded by an annular premixed pilot flame (pilot diameter 15 mm). A low speed annular coflow of filtered air surrounded the burner to eliminate particles from the flow, which would interfere with the Rayleigh scattering. The methane was diluted 3/1 on a volume basis to eliminate soot. The Reynolds number was 20,600 and the beams intersected the flow at 25 nozzle diameters downstream. The signal/noise for the ambient air in the Rayleigh images is  $\approx 200$ . Because of the weakness of the Raman signal, the signal/noise for those images is roughly 15.

Sets of 500 instantaneous shots were taken at a number of different locations within the flame to allow statistical characterization of the mixture fraction and the scalar dissipation. A complete description of the results obtained from this measurement approach has been accepted for presentation at the Twenty Fifth International Symposium on Combustion in Irvine, CA in July 1994. (See publication #28.)

## PUBLICATIONS RESULTING FROM THE RESEARCH

### I. Nonlinear Spectroscopy of Multicomponent Droplets

1. G. Chen, R.K. Chang, S.C. Hill, and P.W. Barber, "Frequency Splitting of Degenerate Spherical Cavity Mode: Stimulated Raman Scattering Spectrum of Deformed Droplets," *Opt. Lett.* **16**, 1269 (1991).
2. R.K. Chang, G. Chen, S.C. Hill, and P.W. Barber, "Nonlinear Optical Processes in Droplets with Single-Mode Laser Excitation", in Proceedings of the SPIE Conference on Nonlinear Optics and Materials, Vol. 1497 (SPIE, Bellingham, Washington, 1991), p. 2.
3. A. Serpengüzel, G. Chen, R.K. Chang, and W.-F. Hsieh, "Heuristic Model for the Growth and Coupling of Nonlinear Processes in Droplets," *JOSA B* **9**, 871 (1992).
4. D.H. Leach, R.K. Chang, and W.P. Acker, "Stimulated Anti-Stokes Raman Scattering in Microdroplets," *Opt. Lett.* **17**, 387 (1992).
5. R.K. Chang and Ali Serpengüzel, "Characteristics and Applications of Stimulated Raman Scattering in Microdroplets," in Thirteenth International Conference on Raman Spectroscopy, 1992, Würzburg, Germany, W. Kiefer, M. Cardona, G. Schaak, F. W. Schneider, and H. W. Schrötter, eds. (John Wiley and Sons, 1992) p. 33.
6. David H. Leach, Richard K. Chang, William P. Acker, and Steven C. Hill, "Third Order Sum Frequency Generation in Droplets: Experimental Results," *JOSA B* **10**, 34 (1993).
7. Steven C. Hill, David H. Leach, and Richard K. Chang, "Third Order Sum Frequency Generation in Droplets: Model with Numerical Results for Third Harmonic Generation," *JOSA B* **10**, 16 (1993).
8. Alfred S. Kwok and Richard K. Chang, "Fluorescence Seeding of Weaker-Gain Raman Modes in Microdroplets: Enhancement of Stimulated Raman Scattering," *Opt. Lett.* **17**, 1262 (1992).
9. J. Christian Swindal, David H. Leach, Richard K. Chang, and Kenneth Young, "Precession of Morphology-Dependent Resonances in Non-Spherical Droplets," *Opt. Lett.* **18**, 191 (1993).
10. Gang Chen, J. Christian Swindal, and Richard K. Chang, "Frequency Splitting and Precession of Cavity Modes of a Droplet Deformed by Inertial Forces," in Proceedings of Shanghai International Symposium on Quantum Optics, Vol. 1726 (SPIE, Bellingham, Washington, 1992) p. 292.
11. Steven C. Hill and Richard K. Chang, "On Modeling Nonlinear Optical Mixing Processes in Droplets," in Proceedings of the SPIE Conference on Laser Applications in Combustion and Combustion Diagnostics, Vol. 1862 (SPIE, Bellingham, Washington, 1993) p. 309.

12. Ali Serpengüzel, Richard K. Chang, William P. Acker, and Rodney L. Sung, "Laser Diagnostic Techniques for Characterizing Droplet Size, Composition, and Differential Evaporation in Fuel Sprays," in XXIV FISITA Congress, 7-11 June, 1992 London, (Mechanical Engineering Publications Ltd, London, 1992). C389/417, paper number 925030, p.107.
13. Gang Chen, Ali Serpengüzel, Richard K. Chang, and William P. Acker, "Relative Evaporation of Droplets in a Segmented Stream Determined by Cavity Droplet Fluorescence Peak Shifts," in Proceedings of the SPIE Conference on Laser Applications in Combustion and Combustion Diagnostics, Vol. 1862 (SPIE, Bellingham, Washington, 1993) p. 200.
14. G. Chen, Md. M. Mazumder, Y. R. Chemla, A. Serpengüzel, R. K. Chang, and S. C. Hill, "Wavelength Variation of Laser Emission Along the Entire Rim of Slightly Deformed Droplets," *Opt. Lett.* **18**, 1993 (1993).
15. Alfred S. Kwok and Richard K. Chang, "Detection of Minority Species in Microdroplets: Enhancement of Stimulated Raman Scattering," *Optics and Photonics News* **4**, 34 (1993).
16. Steven C. Hill and Richard K. Chang, "Nonlinear Optics in Droplets," in Proceedings of the 5th International Topsoe Summer School on Nonlinear Optics (Aalborg, Denmark, 3-8 August, 1992) O. Keller, ed., Nova Science Publishers, New York, (in press).
17. J. Christian Swindal, G. Chen, A. Serpengüzel, R. K. Chang, and W. P. Acker "Spray Diagnostics with Lasing and Stimulated Raman Scattering," Submitted to the AIAA Progress Series "Recent Advances in Spray Combustion," American Institute of Aeronautics and Astronautics, Washington, D.C. (Submitted).

## II. Two- and Three-Dimensional Measurements in Flames

18. J. H. Frank, K. M. Lyons, and M. B. Long, "Technique for Three-Dimensional Measurements of the Time Development of Turbulent Flames," *Opt. Lett.* **16**, 958 (1991).
19. M. B. Long, "Multi-Dimensional Imaging in Combusting Flows by Lorenz-Mie, Rayleigh, and Raman Scattering," in *Instrumentation for Flows with Combustion*, A. K. M. P. Taylor, ed., p. 467-508 (Academic Press, 1993).
20. S. H. Stárner, R.W. Bilger, R. W. Dibble, R. S. Barlow, D. C. Fourquette and M. B. Long, "Joint Planar CH and OH LIF Imaging in Piloted Turbulent Jet Diffusion Flames Near Extinction," *Proceedings of the Twenty-Fourth Symposium (International) on Combustion*, (The Combustion Institute, Pittsburgh, 1992).
21. M. B. Long, S. H. Stárner, and R. W. Bilger, "Differential Diffusion in Jets Using Joint PLIF and Mie Imaging," *Combust. Sci. Tech.* **92**, 209 (1993).

22. R. W. Dibble, A. R. Kerstein, M. B. Long, B. Yip, and K. Lyons, "Measurement and Computation of Differential Molecular Diffusion in a Turbulent Jet," in *Turbulence and Molecular Processes in Combustion*, T. Takeno, ed., p. 303-310 (Elsevier Science Publishers, 1993).
23. J. H. Frank, K. M. Lyons, and M. B. Long, "Simultaneous Scalar/Velocity Field Measurements in Turbulent Gas-Phase Flows," *Proceedings of the Central States Section of the Combustion Institute*, Madison, WI (1994).
24. S. H. Stårner, R. W. Bilger, K. M. Lyons, D. F. Marran, and M. B. Long, "Planar Mixture Fraction Measurements in Turbulent Flames by a Joint Rayleigh and Fuel LIF Method," to appear in *Proceedings of the 14th International Colloquium on the Dynamics of Explosions and Reactive Systems*.
25. M. B. Long, J. H. Frank, K. M. Lyons, D. F. Marran, and S. H. Stårner, "A Technique for Mixture Fraction Imaging in Turbulent Nonpremixed Flames," *Ber. Bunsenges. Phys. Chem.*, **97**, 1555-1559 (1993).
26. P. Lindstedt and M. B. Long, "Comments on the Pyrolysis Properties of Acetaldehyde," Unpublished report (1993).
27. J. H. Frank, K. M. Lyons, D. F. Marran, M. B. Long, S. H. Stårner, and R. W. Bilger, "Mixture Fraction Imaging in Turbulent Nonpremixed Hydrocarbon Flames," accepted for publication *Twenty-Fifth Symposium (International) on Combustion*, Irvine, CA, July 1994.
28. S. H. Stårner, R. W. Bilger, K. M. Lyons, J. H. Frank, and M. B. Long, "Conserved Scalar Measurements in Turbulent Diffusion Flames by a Raman and Rayleigh Ribbon Imaging Method," accepted for publication *Twenty-Fifth Symposium (International) on Combustion*, Irvine, CA, July 1994.

## SCIENTIFIC COLLABORATORS

In addition to the Co-Principal Investigators, the following people have participated in this project:

### I. Nonlinear Spectroscopy of Multicomponent Droplets

Outside Scientists: William P. Acker (Texaco)  
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## LECTURES PRESENTED ABOUT THE RESEARCH

**Richard K. Chang:** (Talks not delivered by Richard Chang are designated by \*).

"Relative Evaporation of Multicomponent Fuel by Stimulated Raman Scattering," Spring 1991 DOE Diesel Group Meeting, Indianapolis, IN, May 2, 1991 (Progress Report).

"Nonlinear Waves in Single Droplets," Optical Engineering Southcentral '91 Meeting on Nonlinear Optics and Materials of the Society for Photography and Image Engineering (SPIE), Dallas, TX, May 8, 1991 (Keynote Lecture).

"Microcavity Characteristics Associated with a Single Liquid Droplet," Joint Meeting of Quantum Electronics and Laser Sciences (QELS) with Conference of Lasers and Electro-Optics (CLEO), Baltimore, MD, May 14, 1991 (Invited Talk).

"Diagnostics in Spray Combustion," 1991 Gordon Research Conference on the Physics and Chemistry of Laser Diagnostics in Combustion, Plymouth, NH, July 15-19, 1991 (Discussion leader of session).

"Optical Diagnostics of Single Droplets," U.S. Army's CRDEC, Aberdeen Proving Ground, MD, September 30, 1991 (Seminar).

"Update of Nonlinear Spectroscopy of Diesel-Fuel Related Droplets," Fall Diesel Working Group Meeting, State College, PA, October 24, 1991 (Progress Report).

"Nonlinear Spectroscopy of Droplets," Annual Meeting of the Optical Society of America (OSA) and Meeting of Lasers and Electro-Optics Society (LEOS), San Jose, CA, November 5, 1991 (Invited Talk).

\* "Frequency Splitting and Precession of Cavity Modes of a Droplet Deformed by Inertial Forces," Shanghai International Symposium of Quantum Optics, Shanghai, China, March 30-April 2, 1992. (Delivered by Gang Chen).

"Nonlinear Optics in Single Droplets," Physics Department Seminar, Harvard University, Cambridge, MA, April 3, 1992.

"Basic Studies and Applications of Nonlinear Emission from Microdroplets," Physics Department, The Chinese University of Hong Kong, Shatin, HK, April 13-15, 1992. (Series of three lectures).

\* "Laser Diagnostic Techniques for Characterizing Droplet Size, Composition, and Differential Evaporation in Fuel Sprays," FISITA Congress, London, England, June 7-11, 1992. (Delivered by William P. Acker).

"Nonlinear Spectroscopy of Multicomponent Droplets," AFOSR Propulsion Contractors Meeting, La Jolla, CA, June 15-19, 1992.

"Stimulated Emission from Microdroplets" The Second Workshop on Quantum Optics, Weizmann Institute of Science, Rehovot, Israel, June 22-26, 1992. (Invited Talk).

\* "Precession of Morphology-Dependent Resonances in Non-Spherical Droplets," (delivered by J. Christian Swindal); and "Fluorescence Seeding of Stimulated Raman Scattering in Droplets," (delivered by Alfred S. Kwok), Scientific Conference on Obscuration and Aerosol Research, U.S. Army Chemical Research, Development and Engineering Center, Aberdeen Proving Ground, MD, June 22-26, 1992.

"Nonlinear Optical Effects in Micrometer Size Particles, I & II," 5th International Topsoe Summer School on Nonlinear Optics, Aalborg, Denmark, August 3-8, 1992. (Two Invited Talks).

\* "Fluorescence Seeding of Stimulated Raman Scattering in Microdroplets," Topical Meeting on the Nonlinear Optics: Materials, Fundamentals, and Applications, Maui, Hawaii, August 17-21, 1992. (Delivered by Alfred S. Kwok).

"Determination of Chemical and Physical Properties of Droplets from Nonlinear Spectroscopy," Edwards Air Force Base, CA, August 21, 1992.

"Characteristics and Applications of Stimulated Raman Scattering in Microdroplets", XIIIth International Conference on Raman Spectroscopy, Würzburg, Germany, August 31- September 4, 1992. (Plenary Lecture).

"Nonlinear Optics inside a Microdroplet," Atomic Physics Seminar, State University of New York, Stony Brook, NY, October 14, 1992.

"Chemical Speciation of a Droplet with Nonlinear Optical Effects," Physics Seminar, RPI, Rensselaer, NY, October 14, 1992.

\* "Third Order Sum Frequency Generation in Droplets," Annual Meeting of the OSA, Albuquerque, NM, September 20-25, 1992. (Delivered by Steven C. Hill).

\* "Status Report of Nonlinear Diagnostics of Droplets and Sprays," UTRC, East Hartford, CT, November 11, 1992. (15 minutes talks delivered by 5 students and 1 post doctoral scientist).

"Evaporation of Droplets as Detected by Stimulated Light Scattering," Air Force Aero-Propulsion Laboratory, Wright Patterson Air Force Base, Dayton OH, December 2, 1992.

"Determination of Chemical and Physical Properties of Droplets from Nonlinear Spectroscopy," NASA Langley, Cleveland, OH, December 3, 1993.

\* "Relative Evaporation Rates of Droplets in a Segmented Stream Determined by Droplet Cavity Fluorescence Peak Shifts," SPIE Conference on Laser Applications in Combustion and Combustion Diagnostics, Los Angeles, CA, January 18-23, 1993. (Delivered by Ali Serpengüzel).

"Optical Diagnostics of Chemical Species in Flowing Microdroplets: Nonlinear Spectroscopy," Chemical Dynamics and Diagnostics Branch, Naval Research Laboratory, Washington D.C., June 21, 1993.

"Nonlinear Optical Processes in a Liquid Droplet Microcavity," 3rd International Conference on Electrical Transport and Optical Properties of Inhomogeneous Media, Guanajuato, Mexico, August 9-13, 1993. (Invited Talk).



**"Physical and Chemical Characterization of a Microdroplet from the Lasing Spectra," Annual Meeting of the Optical Society of America, Toronto, Canada, October 3-8, 1993. (Invited Lecture).**

**"Physical and Chemical Characterization of a Microdroplet from the Nonlinear Optical Spectra," 12th Annual Meeting of the American Association for Aerosol Research, Oak Brook, IL, October 11-15, 1993. (Plenary Lecture).**

### **Marshall B. Long**

**"Digital Imaging of Turbulent Flows," AGARD Consulting Mission to Instituto Superior Tecnico, Lisbon, Portugal, April 28, 1991.**

**"Applications of Advanced Combustion Imaging Diagnostics," MIT Mechanical Engineering Seminar, Cambridge, MA, May 9, 1991.**

**"Two- and Three-Dimensional Measurements in Flames," AFOSR Contractor's Meeting, Boulder, CO, June 10-14, 1991.**

**"Simultaneous Velocity and Scalar Imaging in Flames," Institut fur Technische Mechanik Seminar, RWTH Aachen, Germany, June 25, 1991.**

**"Planar Imaging" presented at The 1991 Gordon Research Conference on the Physics and Chemistry of Laser Diagnostics in Combustion held at Plymouth State College (North) in Plymouth, New Hampshire, July, 1991. (Discussion leader of Session).**

**"Combustion Diagnostics at Yale," The Eighteenth Sandia Turbulence and Aerothermochemistry Research (STAR) meeting held at Yale University, New Haven, CT October, 1991. (Hosted).**

**"Conserved Scalar Measurements in a Turbulent Nonpremixed Flame," The Nineteenth Sandia Turbulence and Aerothermochemistry Research (STAR) meeting held at the Lawrence Berkeley Laboratory, Livermore, CA, April, 1992.**

**"Imaging Measurements in a Combustion Bomb," presented at Texaco, Beacon, New York, January, 1993.**

**"Combustion Diagnostics Using Imaging," Conference on Lasers and Electro-Optics (CLEO '93) Baltimore Convention Center, Baltimore, Maryland, May 2, 1993. (Invited Talk).**

**"Two-and Three-Dimensional Measurements in Flames," AFOSR Annual Meeting Atlantic City, New Jersey, June 14-18, 1993.**

**"Multidimensional Flow Visualization," Meeting on Laser Diagnostics for Industrial Processes, Heidelberg, Germany, June 28 - July 2, 1993. (Invited Talk).**

**"Mixture Fraction Imaging in Turbulent Nonpremixed Flames," presented at The 1993 Gordon Research Conference on the Physics and Chemistry of Laser Diagnostics in Combustion held at Plymouth State College (North) in Plymouth, New Hampshire, July, 1993.**

**"Multi-dimensional Laser Diagnostics of Combustion," Wesleyan University Physics Department Seminar, September 23, 1993.**

**"Mixture Fraction Imaging in Turbulent Nonpremixed Hydrocarbon Flames," Fall Meeting of the Western States Section of the Combustion Institute, Menlo Park, CA, October 18-19, 1993.**

## INTERACTIONS WITH OTHER LABORATORIES

### Richard K. Chang:

Air Force Aero-Propulsion Laboratory, Wright Patterson AFB, OH  
Drs. T. Jackson, M. Roquemore, and Lt. R. Hancock

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Dr. P. Lindstedt

Sandia National Laboratories, Livermore, CA  
Drs. Rob Barlow and Dr. Alan Kerstein

University of Connecticut, Storrs, CT  
Prof. Baki Cetegen

**DEGREES AWARDED**

**Ali Serpengüzel, Ph.D., May 1992. (Thesis advisor, R. K. Chang)**

**Alfred L.S. Kwok, Ph.D., May 1993. (Thesis advisor, R. K. Chang)**

**Jonathan Frank, Master of Science, May 1991. (Advisor, M. B. Long)**

**Kevin M. Lyons, M. Phil., May 1992. (Advisor, M. B. Long)**

**David Marran, Master of Science, May 1993. (Advisor, M. B. Long)**